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Elemental Characteristics of Aerosols Emitted From a Coal-Fired Heating Plant

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SUMMARY

Size-differentiated aerosols were collected downstream from a heating plant fueled with Eastern coal and were analyzed by using the particle-induced X-ray emission (PIXE) technique. Based on aerosol masses collected in various size ranges, the aerosol-size distribution is determined to be trimodal. The three mass peaks are centered at $0.54\text{ }\mu\text{m}$, $4.0\text{ }\mu\text{m}$, and $11.0\text{ }\mu\text{m}$ with relative peak amplitudes of 1.0, 2.1, and 2.5, respectively. The $11.0\text{ }\mu\text{m}$ peak is probably due to the background aerosols present in the surrounding air. Of the various trace elements present in the aerosols, sulphur is the only element that shows very strong concentration in the smallest size group. Iron is strongly concentrated in the $4.0\text{ }\mu\text{m}$ group. Potassium, calcium, and titanium also exhibit a stronger concentration in the $4.0\text{ }\mu\text{m}$ group than in any other group. Lead and bromine, in the relative concentration of 2 to 1, are equally divided between the $0.54\text{ }\mu\text{m}$ and the $4.0\text{ }\mu\text{m}$ groups. Other trace elements - vanadium, chromium, manganese, nickel, copper, and barium - are also equally divided between the $0.54\text{ }\mu\text{m}$ and the $4.0\text{ }\mu\text{m}$ groups. Apparently, all of the trace elements, except sulphur, enter aerosols during the initial formation and subsequent condensation phases in the combustion process. Excess concentration of sulphur in the $0.54\text{ }\mu\text{m}$ group can be accounted for only by recondensation of sulphur vapors on the combustion aerosols already formed and gas-to-particle phase conversion of sulfate vapors at the stack top. Some of the finer aerosols could also be all sulphur. For all trace elements, the $11.0\text{ }\mu\text{m}$ peak is the weakest, indicating that these elements have very low concentration in the background air.

INTRODUCTION

In view of diminishing reserves and rising prices of alternate fossil fuels, coal is becoming an increasingly important fuel to meet the national energy needs. It is replacing oil as the fuel for electric-power generating plants and is under extensive consideration for: (1) coal gasification for the production of H_2 and CH_4 gases, and (2) synthetic fuel production for ground and air transportation. The increasing use of coal as fuel is expected to bring increased environmental pollution with it. The pollution problem is even more severe with Eastern coal due to its higher sulphur content. Even after appropriate cleaning for sulphur removal, the organic sulphur content of Eastern coal is higher than that of Western coal and its use warrants continuous monitoring for its environmental impact. Elemental characteristics of aerosols from various high-temperature combustion sources have been studied for some time at the Langley Research Center. (See refs. 1 to 3.) This report deals with the results of a study of aerosols collected at a local state farm where high-volatile-content bituminous coal from the Western part of the state is used as the fuel for the heating plants. The sulphur and ash contents of the coal - after physical cleaning and scrubbing - were 0.71 and 6.55 percent

by weight, respectively. The heating-plant boilers are of a stoker type, using about 350 kg/hr (800 lb/hr) of coal, and produce heat equivalent to about 185 kW (250 hp).

The aerosols were size differentiated into eight groups ranging from 0.43 μm to 20 μm and were analyzed by the charged particle-induced X-ray emission (PIXE) technique. The results and their implications are discussed herein.

EXPERIMENTAL PROCEDURE

Aerosol-Collection and Analysis

The aerosols were collected about 90 m downwind from the heating plants for a period of 21 hours and 35 minutes by using two calibrated Andersen cascade impactors. Preconditioned Nuclepore substrates were used to collect size-separated aerosols. The substrates were weighed before and after aerosol collection to determine the weights of aerosols collected in various size ranges. The various size ranges and the corresponding aerosol weights are summarized in table I.¹ The aerosol-bearing filters were analyzed for their elemental composition by using the PIXE technique described in references 1 to 3. The concentration of trace elements detected is summarized in table II. (Elements of sodium through phosphorous have not been included in this table.)

Coal-Sample Analysis

The elemental composition of two representative coal samples was also determined. The coal samples were powdered and spread uniformly on Nuclepore substrates for subsequent analysis by the PIXE technique. The results of this analysis are summarized in table III.

DATA ANALYSIS AND DISCUSSION

The aerosol mass-distribution data, summarized in table I, are illustrated in figure 1. In this figure, the aerosol mass distribution is plotted on a log probability graph, where it is expected to be a straight line for a unimodal lognormal distribution. It is obvious from figure 1 that the aerosols are concentrated in three size groups. This multimodal size distribution is further corroborated by the results shown in figure 2 where $\frac{\Delta M}{\Delta \log D_p}$ is plotted as a function of $\log D_p$. Three distinct mass groups centered, respec-

¹In view of the close proximity of the aerosol collection site to the heating plants and the fact that the aerosols were collected only during the intervals when the wind was blowing from the plants toward the aerosol collectors, it is assumed that the aerosols collected were predominantly of the heating-plant origin.

tively, at 0.54 μm , 4.0 μm , and 11.0 μm are seen in the mass distribution of the aerosols.

The experimentally observed mass distribution has first been approximated to a polynomial distribution function of the following form:

$$f(x) = \frac{dM}{dx} = \frac{\Delta M}{\Delta \log D_p} = \sum_{i=1}^{n=7} a_i x^i \quad (1)$$

where

$x = \log D_p$

ΔM aerosol mass on a stage

D_p aerosol diameter

$\Delta \log D_p = \log \left(\frac{\text{Upper 50\% cutoff size}}{\text{Lower 50\% cutoff size}} \right)$ for a stage

The goodness of the polynomial fit to the experimental mass data is verified by comparing the experimental and the calculated aerosol masses, in various ranges, as summarized in table IV.

The least-squares polynomial function of equation (1) was then used to obtain a compound lognormal distribution function of the following type:

$$f(x) = \frac{dM}{dx} = \sum_{i=0}^7 a_i x^i = \sum_{j=1}^3 A_j e^{-\frac{1}{2} \left(\frac{x-x_j}{\sigma_j} \right)^2} \quad (2)$$

where x_j is the peak position for the j th mode ($\log D_{m,j}$), σ_j is the geometric standard deviation for the j th mode $\left(\frac{1}{2.35} \log \frac{D_1}{D_2} \right)$ where D_1 and D_2 are the half-intensity diameters for the j th mode, and $D_{m,j}$ is the median aerosol diameter for the j th peak.

The values of the calculated masses - obtained by using equation (2) - are also summarized in table IV. It is obvious that the calculated masses using the polynomial distribution function and the compound lognormal distribution function are in excellent agreement with each other as well as with the experimentally

observed aerosol masses in various size ranges. In view of this excellent agreement, all subsequent analysis has been conducted in terms of compound log-normal distribution since it graphically illustrates the multimodal nature of the aerosol-size distribution.

As shown in figure 2, the total aerosol mass distribution is trimodal with three distinct groups centered at 0.54 μm , 4.0 μm , and 11.0 μm , respectively. All elemental concentrations, summarized in table II, were also fitted to a trimodal compound lognormal distribution function. The values of the parameters of the three lognormal components for various elements are summarized in table V. Clearly, the peak positions and the peak widths for elemental-compound lognormal distributions are in good agreement, within experimental errors, with the corresponding total mass values. However, the values of the amplitudes of the three modes differ from element to element as well as from the values for the total aerosol mass. It is noted that sulphur is strongly concentrated in the aerosols centered at 0.54 μm , whereas iron is concentrated mainly in the aerosols centered at 4.0 μm . (The difference in the size dependence of concentrations of sulphur and iron in aerosols indicates that not all of the sulphur in coal is of pyritic form.) The 4.0 μm group is also the strongest group in K, Ca, and Ti distributions. Other elements are equally divided between the 0.54 μm and the 4.0 μm groups of the aerosols. It should be also noted that the concentrations of all trace elements are low in the 11.0 μm group of aerosols. This would tend to imply that the 11.0 μm aerosol group is not produced in the combustion zones of the heating plants but is present in the background air. Figures 3(a) and (b) show some typical elemental mass distributions in various aerosol-size ranges.

An alternative approach to the analysis just outlined would be to measure fractional elemental concentration in various aerosol-size ranges. Such fractional concentrations would be expected to illustrate if any elements exhibit preference for residence on the aerosol surfaces. (See ref. 1.) Values of experimental fractional elemental concentrations, $R_{\text{exp}} = \Delta M_{\text{elem}} / \Delta M_{\text{total}}$, for various aerosol-size ranges are summarized in table VI and illustrated in figures 4(a) and 4(b). It is seen that, except for sulphur, none of the other elements show any marked preference for surface residence. Sulphur, on the other hand, seems to be confined strongly to the smaller aerosols - a trend consistent with preference for surface residence. As a matter of fact, R_{exp} for sulphur submicrometer aerosols is in excess of the value calculated on the basis of recondensation of sulphur vapors on the pre-existing combustion aerosol surfaces. (See fig. 4(a).) This excess of sulphur would tend to imply that some of the submicrometer aerosols may be all sulphur or may be the result of heteromolecular nucleation processes (refs. 4 and 5), such as $\text{NH}_3 \cdot \text{H}_2\text{SO}_4 \rightarrow (\text{NH}_4)_2\text{SO}_4$ and $(\text{H}_2\text{O})_n \cdot (\text{H}_2\text{SO}_4)_m \rightarrow \text{H}_2\text{SO}_4$ (dilute), occurring when the hot combustion vapors encounter colder ambient air at the stack top.

Both of the data-analysis techniques - mass-distribution functions and fractional elemental concentrations - suggest that, except for sulphur, all the elements enter the combustion aerosols in the early as well as later stages of their formation. Sulphur, which is concentrated mainly in the smaller aerosols, appears to enter aerosols predominantly by vapor recondensation on preexisting

combustion aerosol surfaces and also, possibly, by heteromolecular nucleation of sulfate vapors at the stack top.

As indicated earlier, two representative coal samples were also analyzed to determine if there were any differences between the aerosol and the coal compositions. The results of this analysis have been summarized in table III. Except for Rb, Sr, Zr, and Ce, all the trace elements present in the coal are also present in the aerosols though their relative concentrations are different. It should, of course, be noted that the relative concentration of various trace elements present in coal does not necessarily have to be the same in the aerosols. This difference may arise from one of the following reasons:

- (1) Selective elemental fractionation in the aerosols
- (2) Escape of some fuel elements in the vapor phase without being incorporated in the aerosols
- (3) Some fuel elements remaining in the bottom ash, without being vaporized or emitted as fly ash

CONCLUSIONS

The following general conclusions have been drawn regarding the aerosols emitted from heating plants fueled with the Eastern coal:

1. Aerosol-size distribution is trimodal - with the three groups being centered at 0.54 μm , 4.0 μm , and 11.0 μm , respectively.
2. The aerosol composition is not exactly the same as that of fuel coal. This is believed to be the result of selective elemental fractionation in the combustion process.
3. Sulphur is very strongly concentrated in the smaller size aerosols - presumably as a result of recondensation of sulphur-compound vapors on the combustion aerosols or heteromolecular nucleation of some sulfate molecules at the stack top. Some of the smaller aerosols may be all sulphur.
4. Iron is concentrated strongly in the 4.0 μm group - presumably as a result of chemical reactions involved during the initial formation of aerosols.
5. The difference in the size dependence of concentration of sulphur and iron in aerosols suggests that not all of the sulphur in coal is of pyritic form.
6. Most of the trace elements - except for S, Fe, K, Ca, and Ti - are roughly equally divided between the 0.54 μm and the 4.0 μm size groups.

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TABLE I.- MASS DISTRIBUTION AS A FUNCTION OF SIZE OF AEROSOLS EMITTED FROM A

COAL-FIRED HEATING PLANT

Stage number	Aerosol-size range, μm	Mass of aerosols collected, a mg	Mean fraction of total aerosol mass	Mean cumulative mass % less than stated stage
0	11 \rightarrow 20	0.980 \pm 0.050	0.264	73.6
1	7 \rightarrow 11	.780 \pm .050	.210	52.6
2	4.7 \rightarrow 7.0	.520 \pm .050	.140	38.6
3	3.3 \rightarrow 4.7	.480 \pm .050	.129	25.7
4	2.1 \rightarrow 3.3	.400 \pm .050	.108	14.9
5	1.1 \rightarrow 2.1	.110 \pm .050	.030	11.9
6	0.65 \rightarrow 1.10	.130 \pm .050	.035	8.4
7	0.43 \rightarrow 0.65	.210 \pm .050	.057	2.7
Backup	\bar{x} 0.43	.10 (approx.)	.027	0.0
Total aerosol mass . .		3.710 \pm 0.450	1.000	100.0

^aThe aerosol collection period was 21 hours and 35 minutes. Thus, at the sampling rate of 0.00047 m³/s, the total value of air sampled was 36.67 m³.

TABLE II.- SUMMARY OF CONCENTRATIONS OF TRACE ELEMENTS PRESENT IN AEROSOLS COLLECTED

AT VARIOUS STAGES OF THE SAMPLING UNITS^a

[Errors are less than 25 percent unless otherwise listed; all loading and
matrix corrections have been applied]

Element	Elemental concentration, ng/m ³ , at stage -								
	0	1	2	3	4	5	6	7	
S	107.6	111.6	103.2	108.1	109.4	138.2 ± 33.4	612.1 ± 73.1	928.4 ± 102.1	
Cl	100.6	104.5	97.1	102.0	102.8	99.6	106.6	101.7	
K	61.2	63.2	72.8 ± 17.5	91.5 ± 19.9	59.2 ± 19.2	54.7 ± 22.8	31.5 ± 23.1	29.5 ± 21.1	
Ca	37.0 ± 14.2	44.1	55.6 ± 11.7	78.2 ± 16.7	28.9 ± 15.2	42.1	45.1	42.1	
Ti	37.8 ± 9.1	14.3 ± 11.2	53.4 ± 12.2	35.1	26.1 ± 11.7	35.1	37.1	35.1	
V	35.0	36.0	33.1	35.1	36.1	34.0	37.1	35.1	
Cr	32.0	33.0	30.1	32.1	32.0	31.0	34.0	32.0	
Mn	29.0	30.0	28.0	29.0	30.0	29.0	31.0	29.0	
Fe	51.4 ± 13.1	33.0	140.8 ± 18.3	232.9 ± 27.0	142.0 ± 19.0	31.0	33.0	26.1 ± 10.8	
Ni	23.0	24.0	22.0	23.0	24.0	21.3 ± 9.3	25.0 ± 10.0	23.0 ± 9.5	
Cu	21.0	22.0	20.0	21.0	22.0	9.7 ± 6.4	22.0 ± 10.0	21.0 ± 10.0	
Zn	27.0	28.0	26.0	28.0 ± 10.0	28.0 ± 10.0	27.0 ± 9.0	29.0	27.0	
Ba	116.2 ± 20.0	119.2	110.4 ± 30.0	116.3	118.3 ± 27.0	49.2 ± 33.8	122.2	115.2 ± 40.0	
Br	49.0	56.0	42.0	46.0	48.0	47.0 ± 11.0	54.0 ± 15.0	48.0 ± 15.0	
Pb	92.0 ± 27.0	105.0	79.0 ± 30.0	87.0	90.0 ± 27.0	89.0	102.0 ± 25.0	91.0 ± 30.0	

^aElements sodium through phosphorous have not been included in this table.

TABLE III.- ELEMENTAL ANALYSIS OF COAL SAMPLES

[Concentrations are expressed relative to Fe content; Fe content of "worst coal" is 2.5 times that of "best coal"]

Element	Relative elemental content (by mass)	
	Best coal	Worst coal
Al	250	400
Si	670	1475
S	585	400
K	120	500
Ca	150	205
Ti	220	305 ^a
V	38	300
Cr	24	300
Mn	≈30	7.8
Fe (reference)	1000	2500
Cu	10.8	29.3
Zn	11.8	24.5
Br	20	≈25
Rb ^a	28	85
Sr ^a	42	70
Zr ^a	≤20	50
Ce ^a	150	145
Pb	≤10	475

^aThese elements, though present in coal samples, were not detected in the aerosols.

TABLE IV.- SUMMARY OF THE EXPERIMENTAL AEROSOL MASS LOADINGS AND THE
CALCULATED MASSES IN VARIOUS AEROSOL-SIZE RANGES

Number	Sampler stage number	$\Delta M_{\text{exp}},^a \text{ mg}$	$\Delta M_{\text{calc}}, \text{ mg}$	
			Polynomial distribution function	Compound lognormal distribution function
1	^b 0	0.980 \pm 0.050	0.796	0.670
2	1	.780 \pm .050	.784	.771
3	2	.520 \pm .050	.536	.580
4	3	.480 \pm .050	.464	.482
5	4	.400 \pm .050	.416	.400
6	5	.110 \pm .050	.142	.154
7	6	.130 \pm .050	.200	.182
8	7	.210 \pm .050	.280	.252

^aAs indicated in the footnote under table I, the total volume of air sampled was 36.67 m³.

^bThe upper limit for stage 0 is uncertain. It has been assumed to be 20 μm for ΔM_{calc} computations.

TABLE V.- SUMMARY OF LOGNORMAL COMPONENT CHARACTERISTICS IN THE SIZE DISTRIBUTION
OF AEROSOLS EMITTED FROM A COAL-FIRED HEATING PLANT

Element	Lognormal component characteristics					
	Group 1 (0.54 μm)		Group 2 (4.0 μm)		Group 3 (11.0 μm)	
	Peak amplitude, $\mu\text{g}/\text{m}^3$	$\log \sigma_1$	Peak amplitude, $\mu\text{g}/\text{m}^3$	$\log \sigma_2$	Peak amplitude, $\mu\text{g}/\text{m}^3$	$\log \sigma_3$
Total mass	40.91	0.1867 ± 0.0100	84.54	0.2054 ± 0.0100	100.90	0.1800 ± 0.0100
S	5.10	0.1867	0.90	0.2054	0.60	0.1742
K	.16	.2146	.60	.2156	.16	.1847
Ca	.24	.2221	.50	.2188	.13	.1919
Ti	.20	.2146	.31	.2106	.15	.1908
V	.20	.2172	.23	.2368	.12	.1952
Cr	.18	.2221	.21	.2287	.10	.1921
Mn	.16	.2146	.19	.2373	.10	.1919
Fe	.15	.2164	1.50	.2263	.20	.1848
Ni	.13	.2221	.17	.2373	.09	.1747
Cu	.12	.2221	.14	.2373	.08	.1796
Ba	.65	.2146	.75	.2156	.40	.1919
Br	.28	.2221	.30	.2373	.20	.1747
Pb	.59	.2221	.59	.2373	.35	.1823
Average geo. std. deviation . . .	0.2164 ± 0.02000		0.2263 ± 0.0200		0.1848 ± 0.0200	

TABLE VI.- RATIO OF ELEMENTAL MASS TO TOTAL MASS FOR VARIOUS ELEMENTS PRESENT IN AEROSOLS EMITTED FROM A COAL-FIRED PLANT

Impactor stage number	Elemental R-values $\times 10^{-3}$													$R_{s,calc}$ ($t = 100 \text{ \AA}$) (a)
	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Cu	Ba	Br	Pb
0	4.03	3.76	2.27	1.39	1.43	1.32	1.21	1.10	1.92 ± 0.05	0.84	0.77	4.36	1.83	3.45
1	5.24	4.91	2.97	2.16	0.66 ± 0.51	1.69	1.54	1.39	1.55	1.14	1.03	5.57	2.64	4.95
2	7.26	6.84	5.13 ± 0.88	3.92 ± 0.84	3.78 ± 0.88	2.35	2.13	1.98	9.93 ± 1.29	1.54	1.39	7.77	2.97	5.57
3	8.25	7.79	6.97 ± 1.54	5.98 ± 1.28	2.68	2.68	2.46	2.20	17.79 ± 1.23	1.76	1.61	8.87	3.67	6.64
4	10.08	9.42	5.43 ± 1.76	2.64 ± 1.39	2.38 ± 1.06	3.30	2.93	2.75	13.02 ± 1.75	2.20	2.02	10.85	4.40	8.25
5	46.02 ± 12.46	33.20	18.22 ± 7.63	14.04 ± 3.80	11.70	11.33	10.30	9.68	10.35	7.11 ± 3.12	3.23 ± 2.13	16.39 ± 11.26	15.66	29.66
6	172.64 ± 20.61	30.07	8.87 ± 6.53	12.72	10.45	10.45	9.61	8.73	9.31	7.11 ± 1.80	6.20 ± 1.60	34.47 ± 10.00	15.22	28.79
7	126.08 ± 17.82	17.76	5.13 ± 3.67	7.33	6.12	6.12	5.57	5.06	4.58 ± 1.89	4.04 ± 1.50	3.67 ± 2.00	20.13 ± 6.00	8.40	15.88
														112.819

^a $R_{s,calc} = \frac{\Delta m_{elem, calc}}{\Delta m_{total}}$ for exclusive residence in a surface layer of thickness $t = 100 \text{ \AA}$.

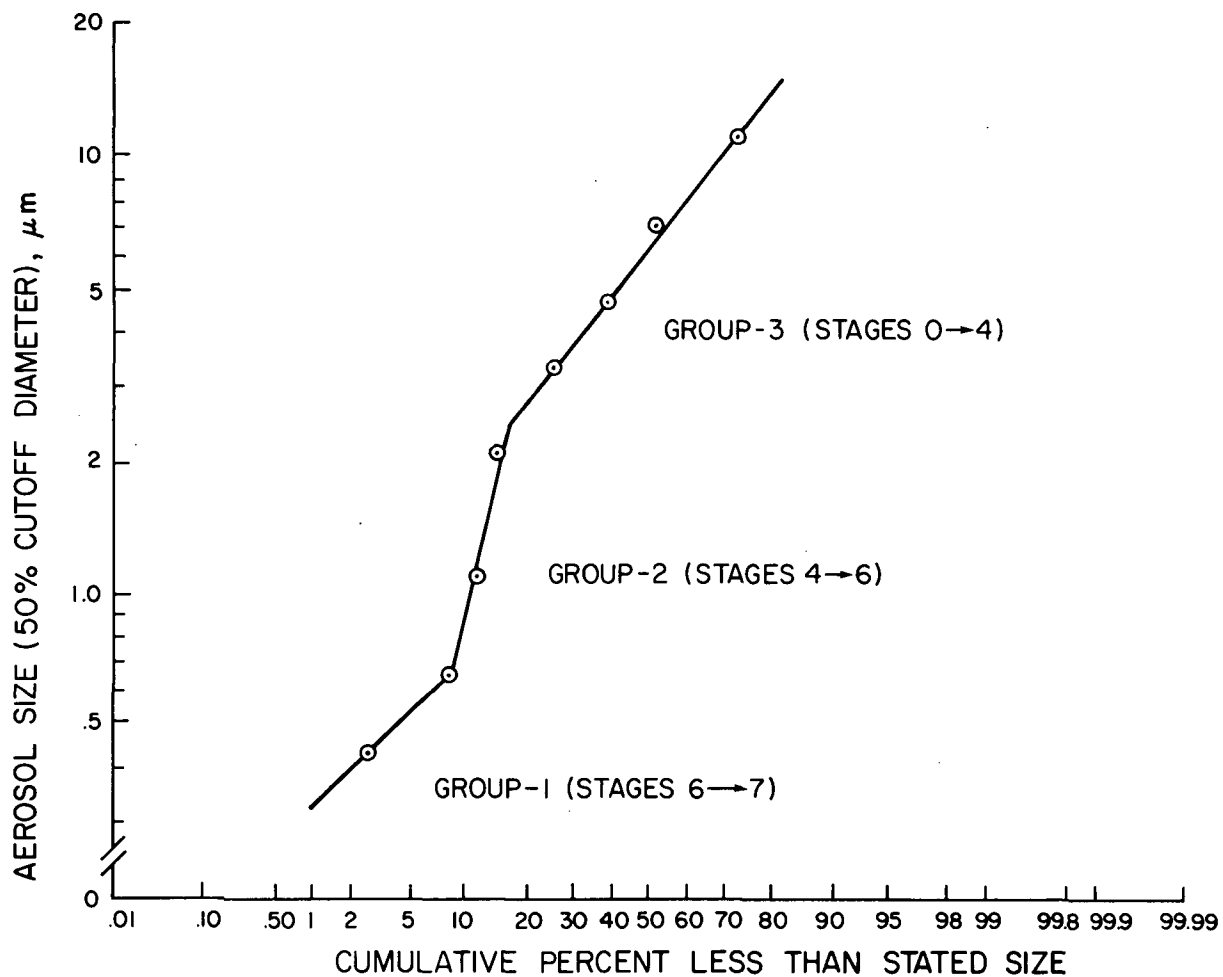


Figure 1.- Mass distribution as a function of the size of aerosols emitted from a coal-fired heating plant.

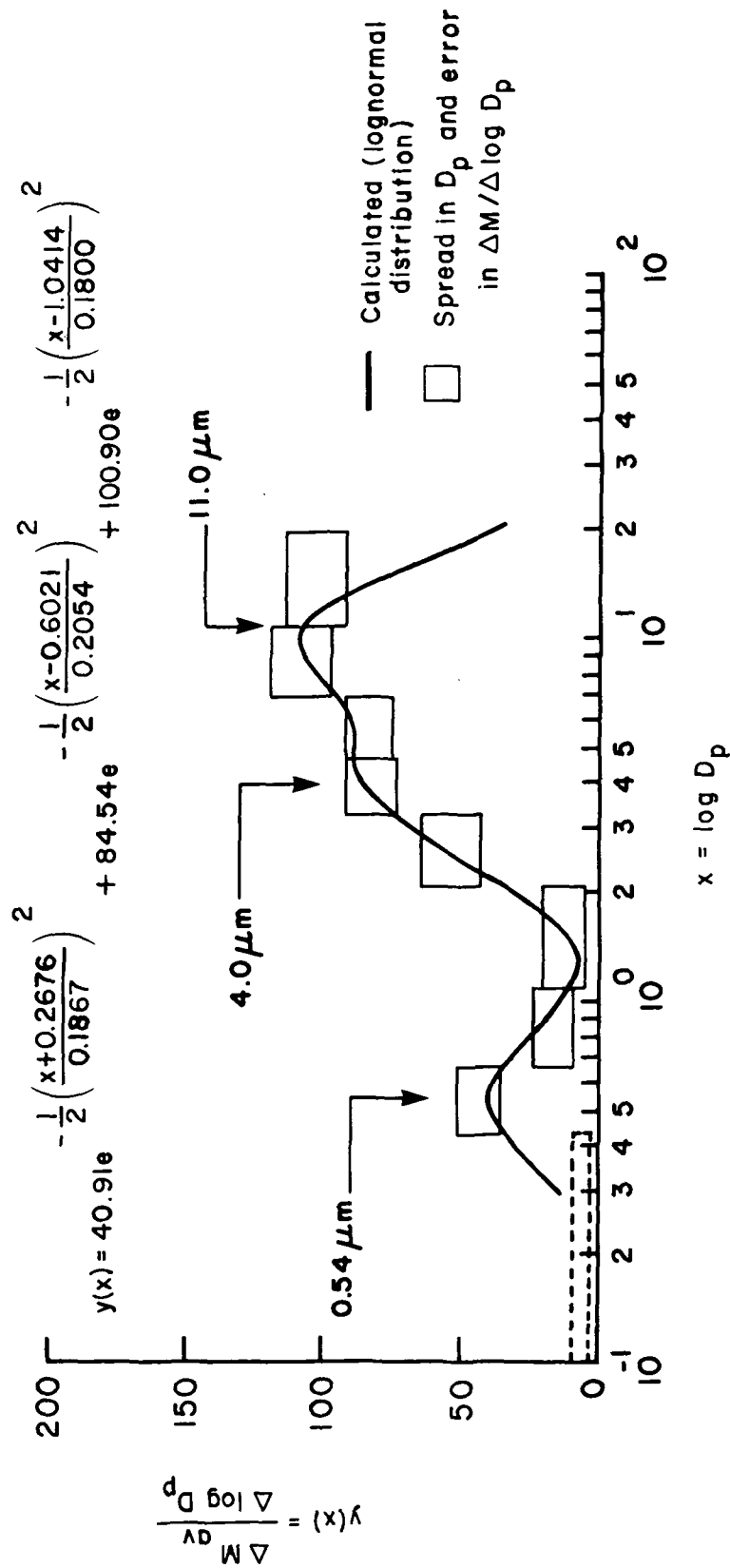
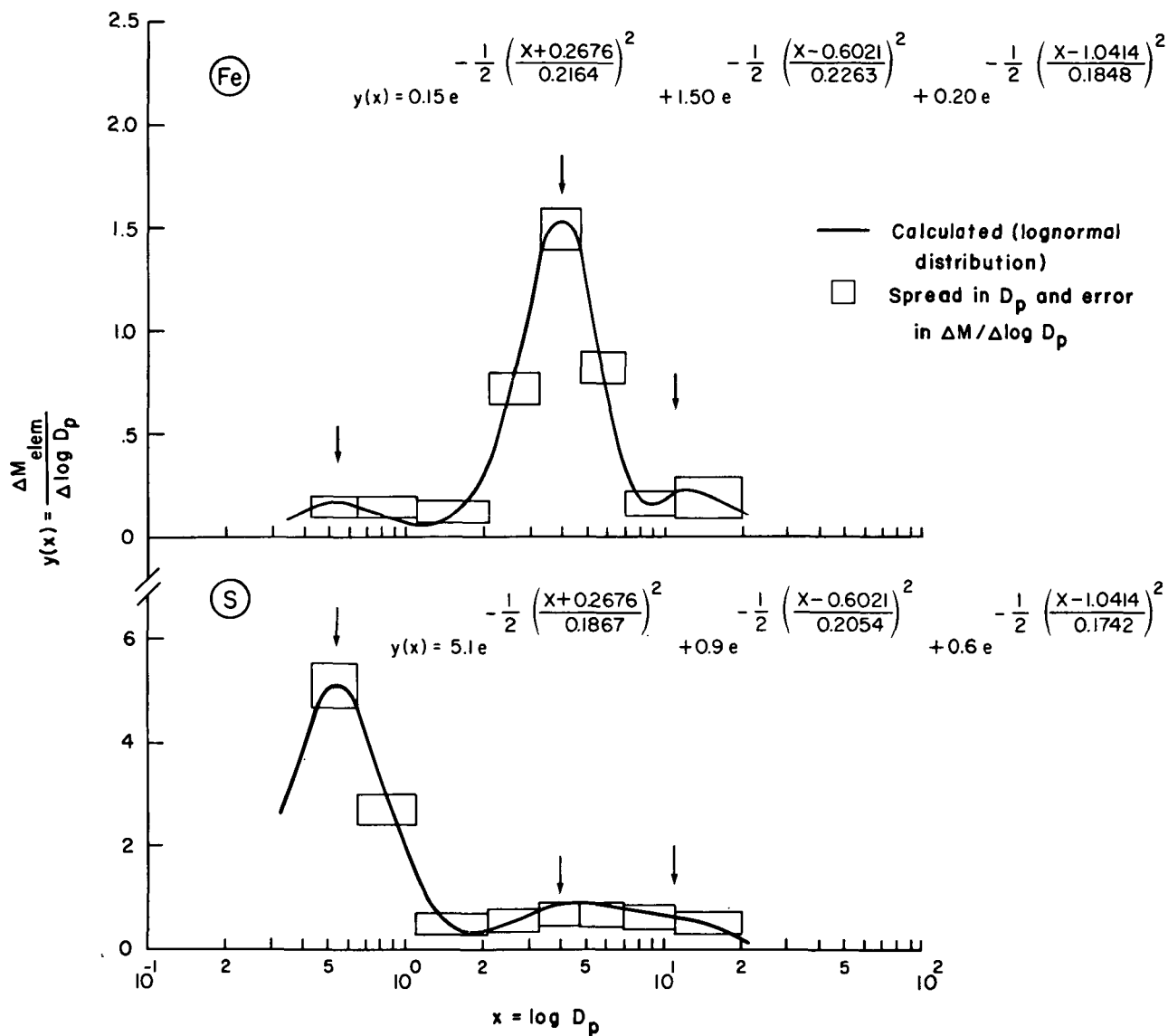
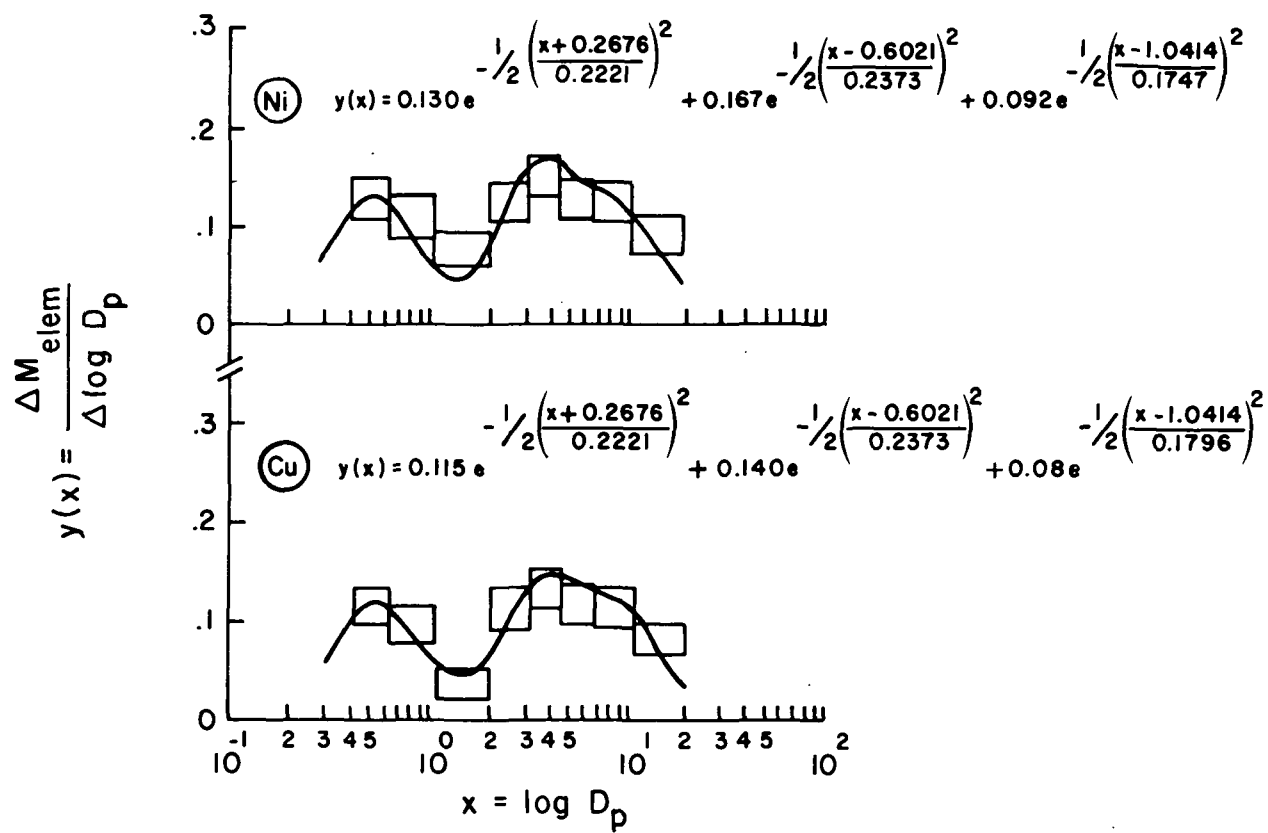


Figure 2.- Total mass as a function of the size of aerosols collected near a coal-fired heating plant. (Average aerosol mass ΔM is in $\mu\text{g}/\text{m}^3$ and the aerosol diameter D_p is in μm .)



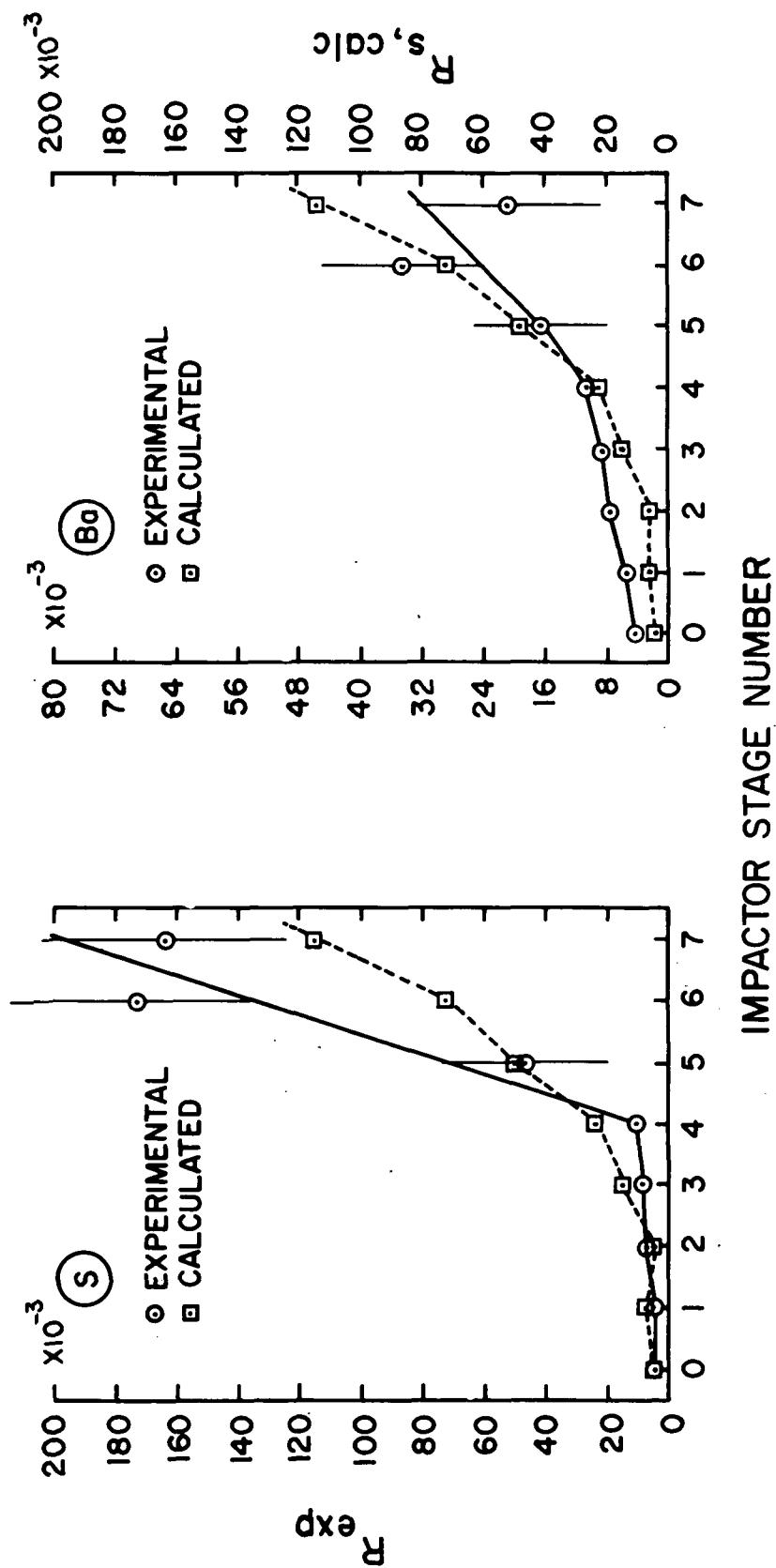
(a) Fe and S.

Figure 3.- Elemental mass as a function of size in aerosols collected near a coal-fired heating plant. (Elemental mass ΔM is in $\mu\text{g}/\text{m}^3$ and particle diameter D_p is in μm .)



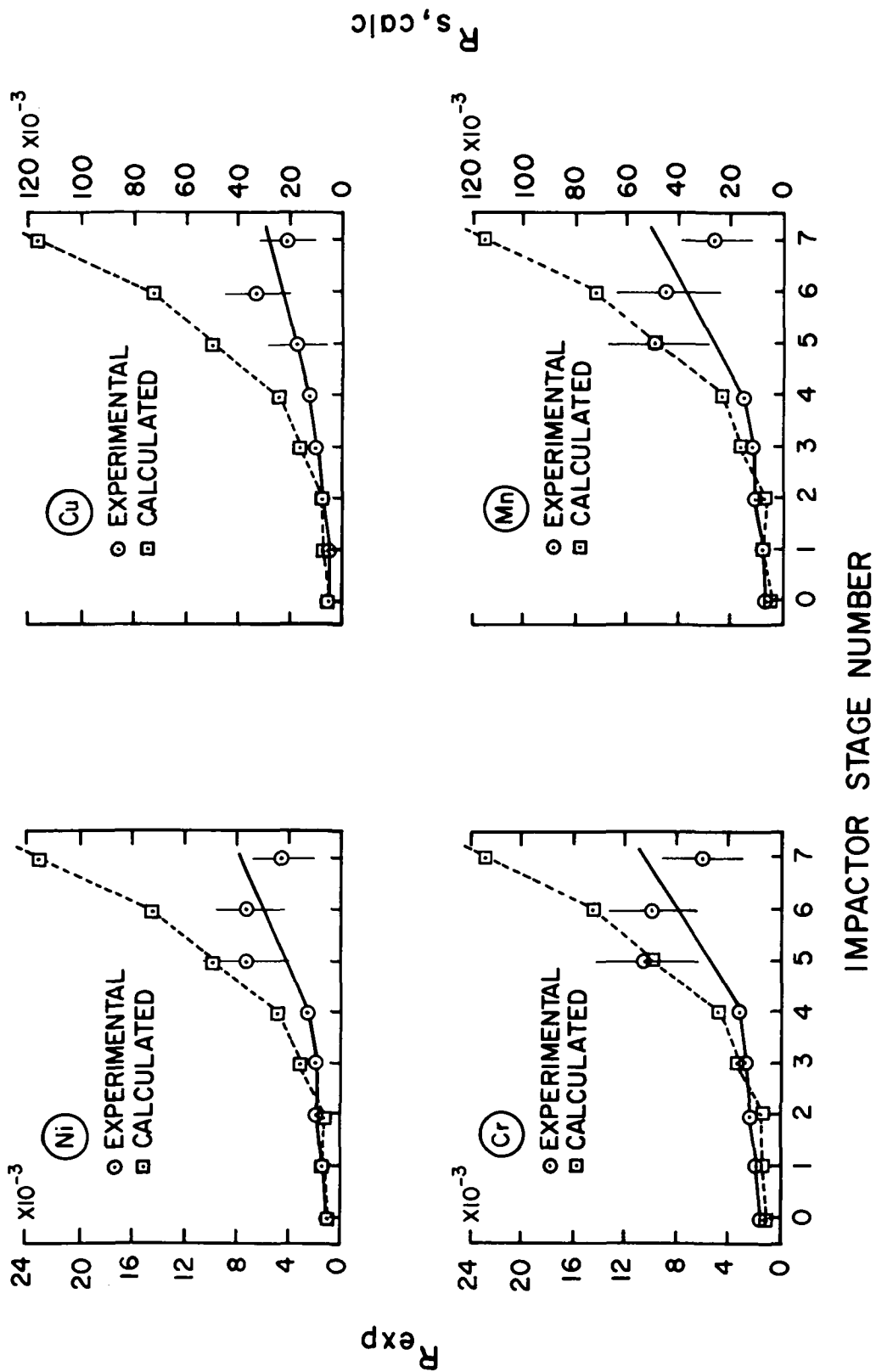
(b) Ni and Cu.

Figure 3.- Concluded.



(a) S and Ba.

Figure 4.- Ratio of elemental mass to total aerosol mass as a function of impactor stage number for aerosols emitted from a coal-fired heating plant.



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